ORIGINAL PAPER

Investigation of contaminant profile in highway stormwater runoff and risk assessment by statistical analysis

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Abstract

Highway stormwater runoff pollution has become a severe risk factor for water bodies nowadays. The conventional risk analysis protocols for directly discharging highway runoff are prone to systematic and judgmental errors. Therefore, a numeric and straightforward risk assessment protocol has been developed in this study that minimizes the errors. For this study, three highway segments were selected in the city of Rawalpindi, Pakistan. Event mean concentrations have been used as baseline numeric values for calculating the risk of discharging highway stormwater directly into the water bodies. These values are also correlated with highway characteristics (area, slope, and traffic count) and storm characteristics (storm depth, cumulative runoff volume, antecedent dry days, and cumulative flow). The highway stormwater was monitored for organics, metals, solids, and macro-nutrients at three highway sections. The event mean concentrations of dissolved organic carbon (50–145 mg/L), total suspended solids (1500–3900 mg/L), chromium (0.25–0.45 mg/L), and lead (0.1–0.8 mg/L) are found to be higher than the environmental quality standards. The risk assessment was conducted by the analysis of variance. The analysis showed that the highway characteristics significantly affect contaminant concentrations, but storm characteristics on contaminant concentrations are not found to be significant. Total suspended solids are the most threatening contaminant in highway runoffs. The study concluded that the risk from contaminants in highway stormwater depends particularly on the specific highway sections' properties. The first flush portion (initial 25% of runoff) of highway runoff poses a higher threat to the receiving environment than the later runoff volumes.

Keywords Risk assessment · Event mean concentrations · Stormwater management · Highway runoff · Urban drainage

Introduction

Highways are one of the primary sources of contaminants to the receiving water environment in an urban environment (Flint and Davis 2007). The highways add a large amount of

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water and, if contaminated, can completely alter the environment of receiving streams (Kriech and Osborn 2022; Calabro 2010). A diversified nature of organic and inorganic contaminants can be found in highway stormwater runoffs (Dang et al. 2022, Huston et al. 2009). On every highway segment, these contaminants need to be controlled and quantified for the sustainability of water and land resources. Among the contaminants, suspended solids are the most abundant contaminant in highway runoffs (Torres 2010). On a highway, vehicles, fuels, and emissions like oil and grease are the source of the primary organic. Highways are also an essential source of metals, mainly from the atmosphere (Dang et al. 2022) and transportation activities (Zafra et al. 2017). The high concentrations of metals in highway runoff have acute and chronic effects on the biotic diversity and marine organisms' mortality rates in the receiving streams (Schiff et al. 2002). The organics and solids facilitate the mobilization of these metals (Nie et al. 2008, Wakida et al. 2014).

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For highway stormwater runoff, risk can be defined as highways' possibility to discharge enough contaminant levels to contaminate the receiving waters. The risk analysis is generally done by allocating scores to the severity level and the likelihood of occurrence (Lundy et al. 2012), which is prone to judgmental errors. Only a risk prioritization protocol can reduce these errors in effective decision-making based on actual numerical values. The contaminant events mean concentrations can give us baseline data to analyze potential risk from the highway stormwater runoffs. Event mean concentrations are the flow-weighted pollutant averages (indices), which can serve as perfect baseline data for selecting, comparing, and implementing the best management practices for stormwater. Event mean concentrations are the single analytical index of the contaminant, which neglects the fluctuations in pollutant concentrations during the storm events and provides an average value. Event mean concentrations are extensively being used to compare multiple storm events and various sites (Chaudhary et al. 2022; Hu et al. 2022; Chow and Yusop 2017). Event mean concentrations represent the pollutant loads at a specific location for a particular storm event. Therefore event mean concentrations are influenced by storm and site characteristics (Kayhanian et al. 2007). The storm characteristics that affect highway runoff contaminant concentrations include storm intensity/depth, duration, and antecedent dry weather. The site characteristics include highway area, slope, and average daily traffic (Liu et al. 2012). During a storm event, the contaminant wash-off is higher in initial runoffs and reduces with increasing runoff volume (Li et al. 2014; Chaudhary et al. 2022).

This phenomenon is called the "first flush". The first flush occurs for two reasons; the contaminant quantity at the source keeps decreasing with increasing runoff, and the increased runoff volume dilutes the available contaminants. These factors work simultaneously to reduce the contaminant concentrations in the later runoff volumes. In a storm management program, the first flush has a significant role (Iqbal and Anwar Baig, 2015). If the first flush is managed, the potential reduction of high loads of contaminants to the receiving waters can be achieved by managing a small fraction of stormwater volume. The objective of the study was to quantify contaminants in highway runoffs. This study conducted a statistical analysis to determine the most influential

factor that affects the event mean concentrations. A risk assessment protocol was devised to analyze the level of risk involved indirectly discharging highway runoff to the water bodies. The risk analysis was performed for the complete storm events and the first flush fraction of those events.

Materials and methods

Highway characteristics

Three highway segments were selected in Rawalpindi, Pakistan, based on traffic count, area, and slopes. Each selected segment had a single drainage point with a high slope ($> 2\%$) to minimize the lag time of stormwater runoff presented in Table 1. Average daily traffic was counted manually according to the method used by Hoeschen et al. (2005). Table 1 shows the characteristics of the three segments. S-III has almost six times bigger than the other two sites with the highest traffic count. S-I was the steepest (slope $= 5$) among the highway segments, with the smallest space and thinnest traffic count.

The average annual rainfall in Rawalpindi, Pakistan, is 990 mm (mainly in July and August), and the temperatures range from $-3\ °C$ to $47\ °C$ (*http*: *//*www.rawalpindi.gov.pk/).

Runoff calculations

An automatic rain gauge (tipping bucket) was installed to measure the precipitation rate at the center of the three sampling sites. The gauge was provided with a data logger, and rainfall was recorded at every 10 min interval. The runoff flow rates were measured for the first three storm events, and the runoff coefficient for each sampling site was calculated (Table 1). The runoff coefficients were then used to calculate the highway discharges for the rest of the storm events by the empirical equation, Eq. 1.

$$Q = CIA \tag{1}$$

where Q is the stormwater flow rate (m^3/hr), C is the runoff coefficient, I is the intensity of rainfall (m/hr), and A is the area of highway segment (m^2).

Sampling

The runoff samples were collected at the drainage points of each highway segment. Samples were collected at 10 min intervals. Sampling was done when runoff started until it completely stopped. Acid-rinsed polyethylene bottles (500 mL) were used for sample collection.

Chemical and statistical analysis

The highway runoff was analyzed for solids, organics, metals, and macro-nutrients. The pH, electrical conductivity,

Table 1 Characteristics of the highway segments

Highway characteristics	Unit	S-I	S-II	S-III
Area	m^2	5300	6900	41500
Slope	%	5	2.4	3
Traffic count	Vehicle/day	25000	25000–50000	50000
Runoff coefficient	–	0.9	0.82	0.87

and total dissolved solids were measured on-site with portable meters (sension + MM150 Portable pH/ORP/EC Multi-Meter, HACH Loveland, CO). The samples were then transported to the laboratory for further physicochemical analysis. The samples were analyzed for Sulphate (Spectronic Genesys 5, spectrophotometer), total phosphorus (acid digested and analyzed with Spectronic Genesys 5, spectrophotometer), total suspended solids (Gravimetric method), chloride (Argentrometry), dissolved organic carbon, and total nitrogen (tot-N, Analytik-Jena, TOC analyzer multi-N/C UV HS, detection limit = 2 µg/L). According to the Standard Methods for the Examination of Water and Wastewater (Carranzo, 2012), all the analysis was done. The samples were digested in HNO_3 before analyzing metals with an atomic absorption spectrophotometer (Hitachi Z-8000, Japan). The lower detection limits for metals were: Cu = 0.3 µg/L; Fe = 0.1 µg/L; Cr = 0.01 µg/L; Pb = 0.03 µg/L, and Zn = 0.01 µg/L.

After the chemical analysis, event mean concentrations were calculated using Eq. 2 (Sansalone and Buchberger 1997)

$$EMC = \frac{M}{V} = \frac{\int_0^{tr} CtQtdt}{\int_0^{tr} Qtdt} \cong \frac{\sum CtQt\Delta t}{\sum Qt\Delta t} \qquad (2)$$

where EMC is event mean concentration (mg/L), M is the total mass of pollutant in the stormwater flow (g), V is the total flow volume of the storm event (m^3), T is time (min), C_t is the concentration at time t (mg/L), Q_t is flow at time t (m^3/min), and Δt is the discrete-time interval (min).

The storm and highway characteristics were correlated with the contaminant event mean concentrations with linear regression. The analysis of variance (ANOVA) was done, and highway segments were compared by Tukey's test. The calculations and statistics were done with the R package (Team, 2013).

Results and discussion

Runoff quantity

Figure 1 shows the characteristics of the nine storm events monitored. The storm depth and duration of the storm events are shown in Fig. 1a, and the resulting runoffs are shown in Fig. 1b. Four of the storms occurred in summer and five during the winter months. Event 2 had the highest storm depth in a comparatively shorter period, and event 8 was the longest storm event (Fig. 1a). We get storm intensity if we divide storm depth by event duration (Serrano). Event 2 and 8 had the highest and lowest intensities among the observed storm events. The total runoff produced by event 2 is almost three times higher than event 8. The antecedent dry days between the storm events ranged from 10 to 31 days. The longest dry days (31 days) were between
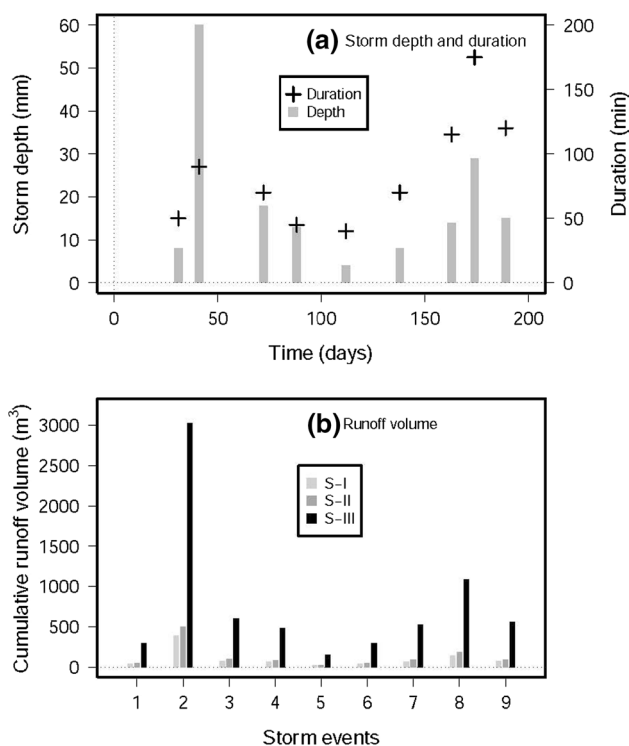

Fig. 1 Characteristics of the storm events; depth and duration of the storm events over the observed period **a**, and the resulting cumulative runoff volumes in each storm event **b**

event 2 and event 3. Figure 1b shows that S-III produced higher runoff volumes (100–3000 m^3) than the other two segments due to the larger area.

Runoff quality

The non-point sources of contaminants include organic and inorganic substances leaked on the highways. The organics include oils, greases, and gasoline from vehicles, and inorganic comes from vehicular exhaust, worn tires, engine parts, brake linings, weathered paint, galvanized vehicle part, and rust. For highways, the contaminant concentrations are mainly dependent on traffic activities. Still, the rainfall (intensity and duration) and catchment (area, slope, and surrounding land use) characteristics cannot be ignored (Opher and Friedler 2010).

Pollutographs

The storm's intensity is one of the most critical factors affecting the contaminant wash-off, especially on a paved area. Figure 2 shows the comparison of chemical oxygen demand of the runoff produced at the three sampling sites between high (event 2) and low (event 8) storm intensity events. Figure 2 shows that

S-III produced a comparatively higher runoff volume than the other segments, but the initial concentrations of chemical oxygen demand were lower than the other two sites. The initial high concentration of chemical oxygen demand at S-I indicates the dilution effect of high runoff volumes. The quick drop in chemical oxygen demand represents a first flush phenomenon. The first flush phenomenon is more pronounced in high-intensity storm events than inmore pronounced high-intensity storm events than in low-intensity events. The chemical oxygen demand fluctuated throughout the low-intensity storm events.

The high contaminant concentrations in highway runoff also depend on the amount of contaminants present on the highway segment. The amount of contaminants on the highway is a function of traffic activities, highway area, and antecedent dry weather period (Maniquiz et al. 2010). Figure 2 suggests that the storm intensity plays a vital role in the quick discharge of contaminants, but other site-specific characteristics cannot be neglected either.

Contaminant event mean concentrations

The event mean concentration is volume-based average contaminant concentration that reduce the fluctuations in runoff volume and represent a mean contaminant value for a particular storm event. Figure 3 shows the event mean concentrations calculated for each contaminant at the three highway segments. The average event mean concentrations for pH, metals, solids, organics, and macro-nutrients are compared with the national environmental quality standards.

The pH of raindrops before hitting the ground is 5.6 (Serrano) due to acidification by CO_2, SO_2, and NO_2 in the atmosphere. The pH of highway runoff was near neutral (pH = 7). This shows that the contaminants on the highway surface neutralized the pH of direct rain. The low pH rainwater has SO_4, which corrodes heavy metals and, as a result, neutralizes the stormwater (Ahmadi et al. 2018). The pH of highway runoffs at each site remained within the environmental quality standards (Fig. 3). The Fe, Cu, and Zn

Fig. 2 Polluto-and hydro-graphs of chemical oxygen demand in the highway stormwater runoff produced by low and high-intensity storm events at three highway segments
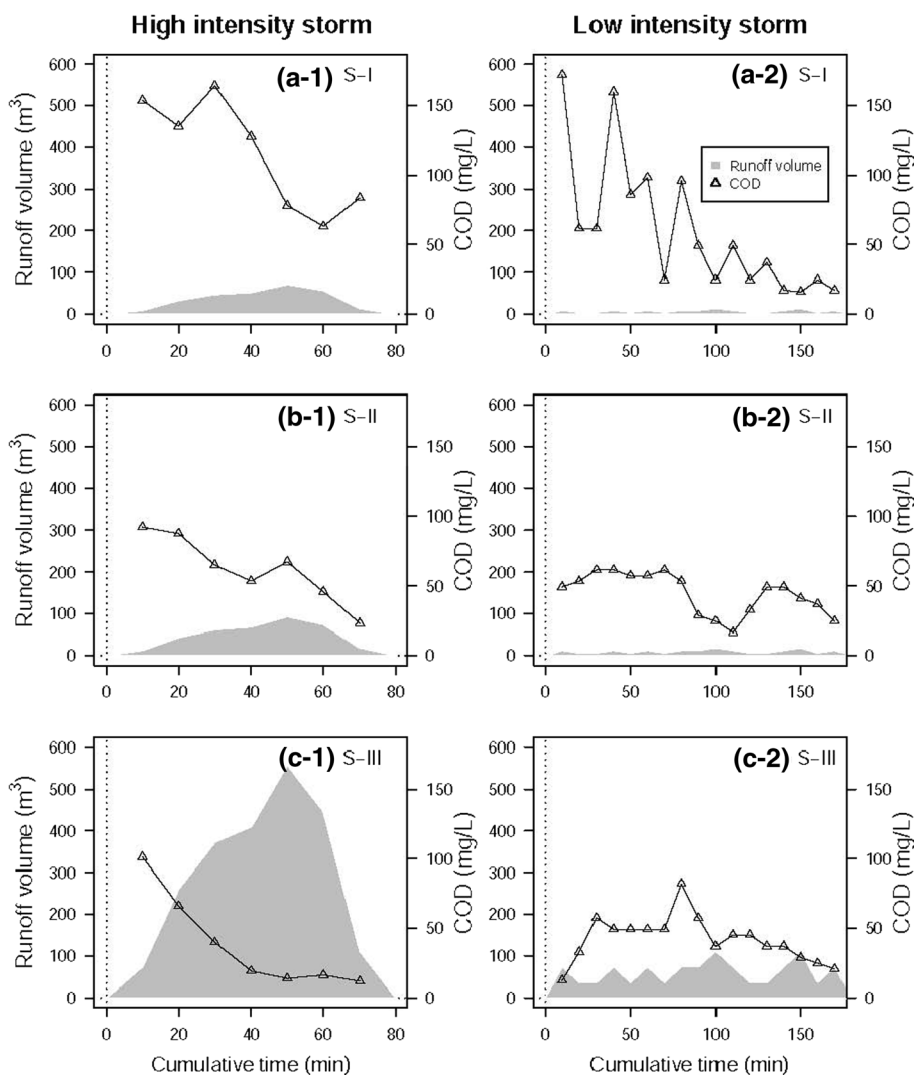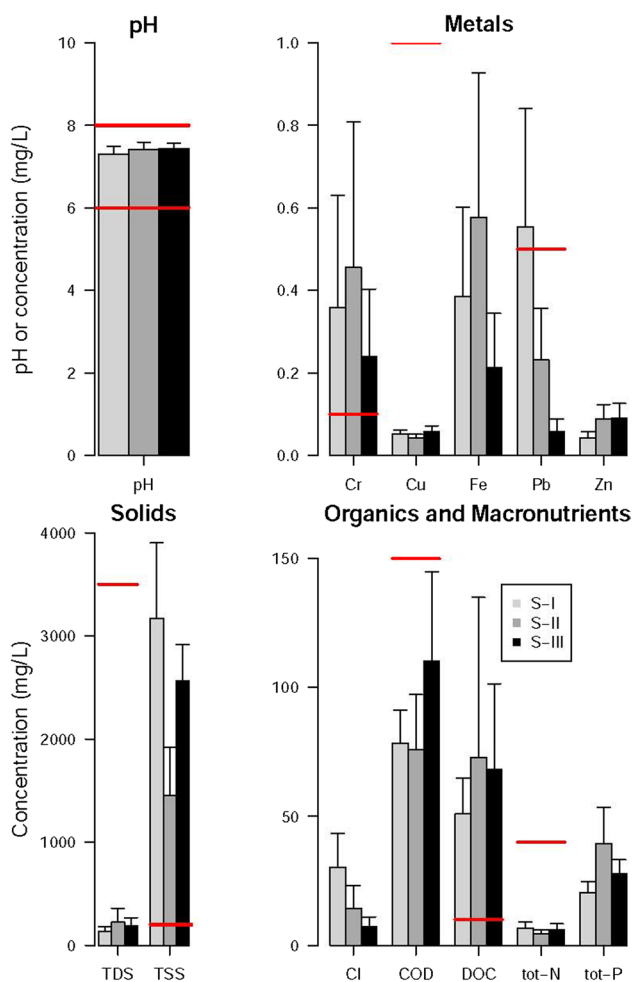
Fig. 3 The contaminant event mean concentrations in the stormwater runoff produced by the three highway segments. The red lines indicate Pakistan's national environmental quality standards for wastewater discharge. Note that there is no standard limit for tot-P, whereas Cl^- and Zn have a limit of 1000 mg/L and 5 mg/L, respectively, but the red lines have fallen out of the plot area. Two red lines for pH show the range of allowed limits

Fig. 4 Boxplots of event mean concentrations of dissolved organic carbon, total suspended solids, chromium, and lead at three highway segments. Asterisks and plus symbols show the significant difference ($p = 0.05$) between the two highway segments

zone habitat (Chapman et al. 2017). Various studies have reported the toxicity due to metals-laden suspended solid discharged into the streams (Ahmadi et al. 2018). Chloride, chemical oxygen demand, and tot-N remained within the environmental quality standards at all the highway segments. The event mean concentrations of dissolved organic carbon were almost five times higher than the highway segments' environmental quality standards.

Effect of highway characteristics on event mean concentrations

The event mean concentrations of contaminants higher than the environmental quality standards at the three highway segments are compared in Fig. 4. There was no significant linear correlation between the catchment characteristics and event mean concentrations, but the general trend of the relationship can be drawn from Fig. 4. The dissolved organic carbon concentrations decreased with the increasing slope of the highway segment. Dissolved organic carbon was highest at S-II, which had the lowest highway slope, and the values were significantly higher than S-I, which had a steeper slope. Total suspended solids concentrations were almost a mirror image of dissolved organic carbon, as the concentrations were higher in high slope segments. Chromium was not significantly different at all sites. Lead concentrations decreased with increasing segment area and traffic count. Kayhanian et al. (2003) compared the event mean concentrations with traffic count and found no significant correlation, whereas Li et al. (2014) found a positive correlation.

among dissolved metals remained within the environmental quality standards. The event mean concentration of Pb at S-I runoff was higher than the environmental quality standards. Most of the time Pb concentrations are attributed to atmospheric deposition, whereas Cu and Zn are sourced from vehicle brake emissions and tire wear, respectively (Rasmussen et al. 2013). Chromium was the only metal that remained higher than the environmental quality standards at all the sites. Li et al. (2014) outlined the potential sources of metals in urban pavement runoff and suggested tire wear as the only primary chromium source. The dissolved solids were within the environmental quality standards, whereas event mean concentrations of total suspended solids were 20 times higher than the environmental quality standards. These high suspended solid concentrations increase the mortality rate of flora and fauna in the receiving stream by physically covering the benthic
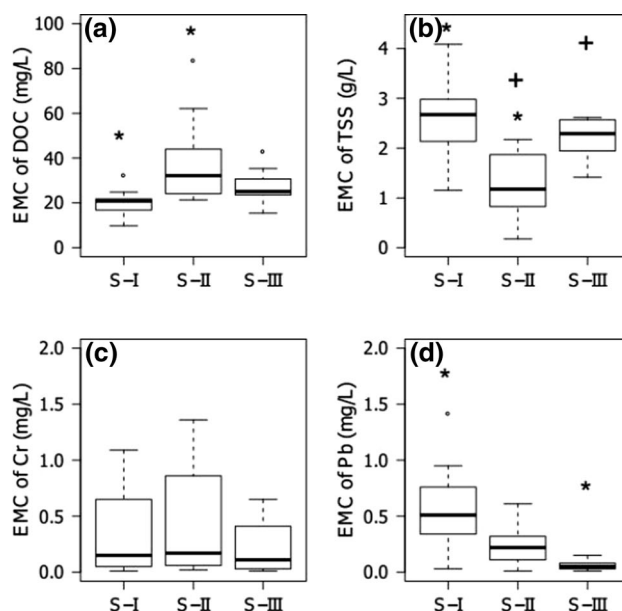
The event means concentrations depend on the specific area under observation. The general trend of contaminant event mean concentrations in Fig. 4 shows that the contaminant concentrations were not affected significantly by the highway characteristics. Combining these factors (contaminant properties, slope, traffic count, and catchment area) collectively participates in high contaminant concentrations.

## Effect of storm characteristics on event mean concentrations

To observe the effect of storm characteristics on event mean concentrations, event mean concentrations of lead were plotted against storm depth, total runoff volume, antecedent dry days, and cumulative flow (Fig. 4). There is no significant correlation between the event mean concentrations and storm characteristics, as suggested by Opher and Friedler (2010). Still, the trend of event mean concentrations influenced by individual factors can be observed. Lead concentrations decreased with increasing storm depth, cumulative runoff volume, and cumulative flow (Fig. 5a, b, d). High storm depth produces high flow and volume, possibly diluting lead

concentrations. Smith et al. (2002) suggested that storm depth is the most crucial factor to be considered when the first flush is needed to be separated from the total runoff volume. Figure 5c shows the antecedent dry days between the storm events increased the event mean concentrations of lead in highway runoff. The contaminants accumulate on the highways during dry days, and storm events mobilize those deposited contaminants (Li et al. 2014). Therefore, high amounts of contaminants are expected when there is a more extended antecedent dry period and vice versa.

Risk assessment

A related study reported that roads and airports have acute and chronic effects on the biotic diversity and marine organisms' mortality rates (Hvitved-Jacobsen et al. 2010). The ecological damage depends on the nature and quantity of the contaminant (Karlsson et al. 2010). The environmental quality standards serve as a baseline for water and wastewater flows and discharges. The risk assessment and prioritization are the most crucial decision-making component in selecting the best management practices for highway stormwater runoff. Risk is the probability of a consequence due to exposure to a physical, chemical, or biological

Fig. 5 The event mean concentrations plotted against storm characteristics; storm depth **a**, total runof volume **b**, antecedent dry days **c**, and cumulative flow **d**. The trend line and regression coefficient (R^2) is added in each plot
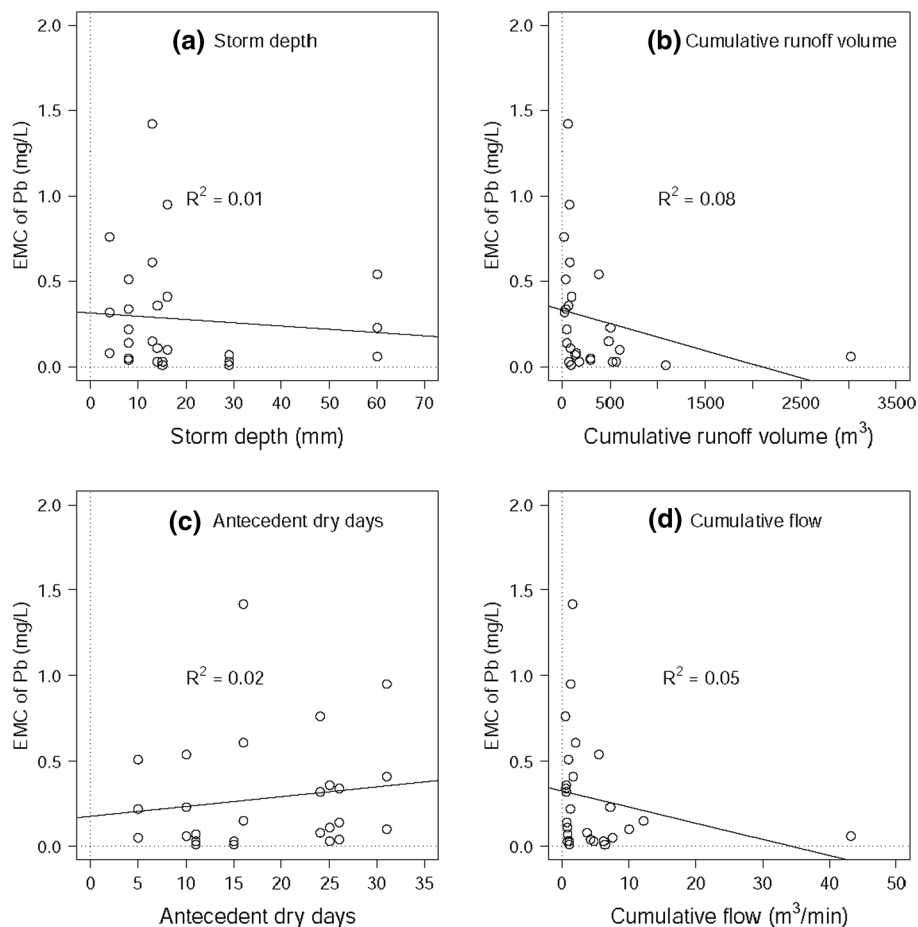
severity. The quality of the risk assessment assures the success and sustainability of best management practices. The risk assessment requires a reference that can be used for comparing the actual field data. Lundy et al. (2012) used environmental quality standards as the reference and event mean concentrations as field data. Risk can be calculated if we know the severity and occurrence of stress. The water quality index is the ratio of contaminant concentrations and environmental quality standards for a particular contaminant (Smith et al. 2002). Therefore, the water quality index can be used as the measurement of severity for a specific contaminant. The occurrence of severity can be a ratio of the number of events that a contaminant event mean concentration remained higher than the environmental quality standards. The product of occurrence and severity of a contaminant in highway runoff can then be scaled as low, moderate, and increased risk. The risk factor was determined through Eqs. 3, 4 and 5.

$$R = SO \tag{3}$$

such that,

$$S = \frac{\sum EMC}{EQS(N)} \tag{4}$$

$$O = \frac{n}{N} \tag{5}$$

where R is the risk, S is the fraction of severity levels, O is the occurrence of the severity, EMC is contaminant event mean concentration, EQS is the environmental quality standard for that contaminant, n is the number of events in which event mean concentrations breached environmental quality standards, and N is the total number of events observed. The higher event mean concentrations and frequent occurrences will lead to higher risk and vice versa.

The scale of risk is shown in Fig. 6. The figure shows risk analysis for a few parameters with high concentrations in highway runoff. Total suspended solids posed a high risk at S-I and S-III and a moderate risk at S-II (Fig. 6a). The dissolved organic carbon and Cr remained at moderate risk at S-II, whereas low risk at S-I and S-III. The rest of the contaminants remained in the quiet risk zone. Figure 6b shows risk analysis for the partial event mean concentrations for the first flush (initial 25% of the storm runoff). The first flush contained higher contaminant levels and hence posed a higher risk to the receiving water body. The suspended solids posed a high risk at all the sites for the first flush. The dissolved organic carbon showed moderate risk at S-II and S-III, and Cr posed moderate risk at S-I and S-II. The runoff volume diluted these contaminants after the first flush, which can be observed by comparing Fig. 6a and Fig. 6b. When later, less contaminated runoff reaches the receiving water body, the first flush could have already deteriorated the quality of the water body.
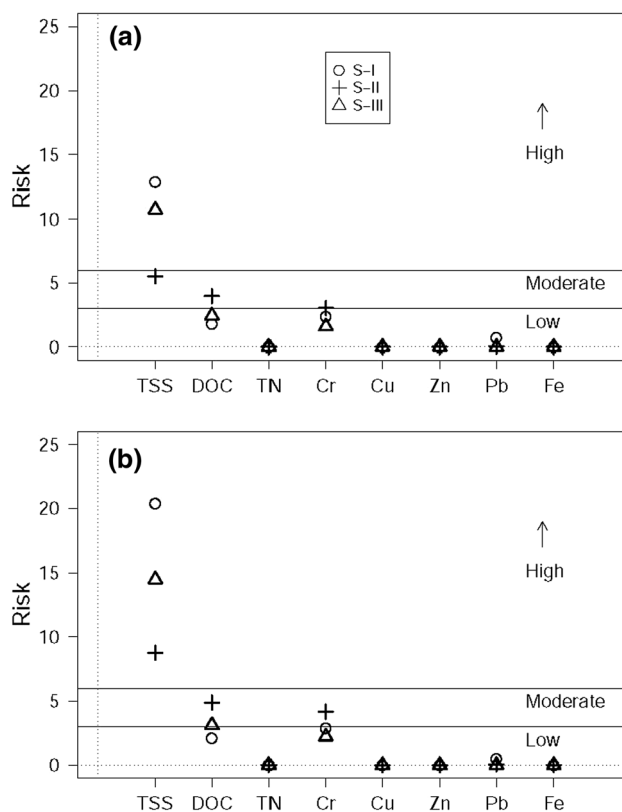

Fig. 6 Risk analysis of the contaminants in highway runoffs at three highway segments. Figure **a** shows the risk analysis based on total EMC, and Figure **b** represents the risk analysis based on Partial EMCs at first flush (Initial 25% of storm runoff). Plot areas are divided into three risk levels (labeled on the right side of the plot), low, moderate, and high risk. The arrow shows that all the values that are higher than ten fall in the high-risk zone

The risk analysis method is quick, simple, numeric, and easy to use. The acceptance levels (low, moderate, and high) of risk can be changed according to the decision maker's need, ease, and type or cost of best management practices.

Conclusion

The contaminant concentrations were higher in the initial highway runoff and decreased with increasing cumulative flux. The decreasing trend of contaminant concentrations was well pronounced in high-intensity storms. The event mean concentrations of dissolved organic carbon (50–145 mg/L), total suspended solids (1500–3900 mg/L), chromium, and lead (0.1–0.8 mg/L) are found to be higher than the environmental quality standards. The highway characteristics significantly affect contaminant concentrations, but storm characteristics on contaminant concentrations are not found to be significant. Total suspended solids are found to be the most threatening parameter in highway runoffs. The study concludes that the risk from contaminants in highway

stormwater depends, particularly on the specific highway sections' properties. The first flush portion (the first 25% of runoff) of highway runoff poses a higher threat to the receiving environment than the later runoff volumes.

Data availability The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declarations

Conflict of interests The authors declare no conflict of interest.

Consent for publication We undertake and agree that the manuscript submitted to your journal has not been published elsewhere and has not been simultaneously submitted to other journals.

Ethics approval The facts and views in the manuscript are solely ours, and we are responsible for authenticity, validity, and originality. We also declare that this manuscript is our original work, and we have not copied it from anywhere else. There is no plagiarism in my manuscript.

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